Parity-Breaking Phase Transition in Tangentially Anchored Nematic Drops (*).

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Summary. — The existence of the parity-breaking phase transition between untwisted and twisted bipolar structures of the tangentially anchored spherical nematic drops has been shown experimentally. Twisted structures behave like the optically active objects in spite of the nonactivity of nematic and isotropic matrix themselves. Temperature dependences of the order parameter (twist angle) have been determined for the drops of 8CB and 8OCB. It has been shown that these dependences are very similar to that observed in usual second-order phase transition.

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Liquid crystalline drops, freely suspended in isotropic fluid or polymer, possess many unusual phenomena in comparison with the drops of isotropic liquids. For example, there are orientational interactions of one drop with the other [1, 2] or with applied field [3, 4], ability of spontaneous division [5], etc. All these phenomena are caused by the ordered structure of the liquid crystalline drops. The order parameter configuration in the drop depends on the type of liquid crystal, elastic constants, surface anchoring, etc. In the simplest case of the nematic drop with tangential boundary conditions, bipolar structures of director have been observed, see, *e.g.*, ref. [6]. These structures contain only two types of deformations: bend and splay, fig. 1a. More recently, however, it has been shown for some types of nematic substances that the bipolar structures can contain twist deformations [7], fig. 1b), c).

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Fig. 1. – Untwisted (a)) and twisted (b), c)) structures of the tangentially anchored nematic drops; a), b) director distributions on the drop surface, c) intersection of twisted structure with vertical plane.

Later Williams [8] has theoretically demonstrated a second-order parity-breaking phase transition between above-mentioned untwisted structure and twisted one. The existence of similar transitions has been demonstrated for normally anchored drops with point defects of hedgehog type [9]. However, the behaviour of the order parameter of transition has not been studied. The purpose of the present communication was to investigate experimentally the possibility of parity-breaking phase transition in bipolar drops and to study the behaviour of the order parameter (twist angle) during this transition.

As follows from the theory [8], untwisted drop is realized when

(1)
$$f(K_{ii}) = K_{33} - 2.32(K_{11} - K_{22}) > 0,$$

where K_{11} , K_{22} and K_{33} are the splay, twist and bend elastic constants, respectively. For $f(K_{ii}) < 0$ the twisted configuration is the minimum energy. Suitable changes of $f(K_{ii})$ sign with temperature are possessed by substances with nematic and smectic phases. Two substances were chosen for investigation: 4-n-octyl-4'-cyanobiphenyl (8CB) and 4-n-octyloxy-4'-cyanobiphenyl (8OCB). The parts of the phase diagrams of interest to us can be described as follows:

$$\frac{8 \text{ CB}}{8 \text{ OCB}} \text{ smectic } A \frac{32.5}{67} \text{ nematic } \frac{40 \text{ }^\circ \text{C}}{80 \text{ }^\circ \text{C}} \text{ isotropic liquid.}$$

Dependences of $f(K_{ii})$ on temperature for 8 CB and 8 OCB are shown by dashed lines in fig. 2a), c); experimental data or ref. [10] were used for the calculations of $f(K_{ii})$.

Spherical drops of tangentially anchored nematic were created by dispersing a liquid crystal in glycerine isotropic matrix [7]. The radius of these drops was $20 \,\mu$ m. A sample inside a quartz or glass cell was placed in a heater where temperatures were kept constant within 0.02 °C. The temperature was varied at the rate of 0.03 °C/min. The drop textures were examined using a PERAVAL Interphako microscope (made by Karl Zeiss, Jena, DDR) modified by attachments for observations in polarized light. The drop axes under investigations were oriented in the horizontal plane along the polarization direction of one of the Nicols crossed at right, angle.

Two types of nematic drop textures were observed. In the first-type texture, the central part of the drop is extinguished, *i.e.* the projections of the director lines into the plane defined by the crossed Nicols are parallel to one of the polarizers. To choose the real direction of these lines a quartz wedge was used and the observations, similar to those described early[7], led us to the conclusion that the lines of the director are parallel to meridional curves which join two diametrically opposite singular points on the drop surface, fig. 1a).



Fig. 2. – Twist angle z_m as a function of temperature for 8CB (a), b)) and 8OCB (c), d)). Dashed curves: linear combination of elastic constants $f(K_n)$ as a function of temperature for 8CB (a)) and 8OCB (c)).

In the second-type texture the extinction is absent when Nicols are crossed at right angle. The extinction of the central part of the drop occurs only when the Nicols are turned through some relative angle $\gamma \neq \pi/2$. These drops behave as optically active objects. The wave guide effect attests the presence of the twist deformations inside the drop, fig. 1b), c). Connected with γ is the angle $2\alpha_m$ of rotation of the polarization plane of the beam passing through the drop, as well as the angle $2\alpha_m$ between the director lines in the lower and upper hemispheres of the drop surface:

$$(2) \qquad \qquad 2\alpha_m = \pi/2 - \gamma$$

From the last relation we have determined the angle α_m as a function of temperature, fig. 2, 3. It should be emphasized that relation (2) is based on the assumptions that a) the twist relaxation is uniform throughout the drop and the angle α_m between the director lines and planes of constant azimuth monotonically decreases from $\alpha = \alpha_m$ at drop surface to $\alpha = 0$ near the axis, b) the Mauguin limit for waveguide regime [11] is satisfied.

As follows from the theory [8], assumption a) is reasonable when $K_{33}/K_{11} \leq 4/3$. The last condition is really satisfied for 8 CB and 8 OCB in the temperature range where the twisted structures exist, see the experimental data of ref. [10]. Assumption b) is reasonable too: a spatial period of the twist distortion is much larger than the optical wavelength.

As can be seen from fig. 2a)-d), the nematic drops of both substances undergo the parity-breaking transformations. It is important to note that in 80CB the untwisted phase possesses a reentrant behaviour. All parity-breaking transitions are reversible

and are separated unambiguously from the points of smectic A-nematic and nematicisotropic liquid transitions. The locations of the parity-breaking transition points T_c is in a good agreement with the predictions of the theory, *i.e.* with the locations of the sign changes of $f(K_u)$.

The available experimental evidence shows that $\alpha_m vs$. temperature curves are very similar to that observed for the order parameter in usual second-order phase transitions. In the vicinity of the points T_e the angle α_m is expected to behave as

$$\alpha_m \sim (T-T_c)^3,$$

where β is the critical index. To obtain β the data were plotted as logarithms of α_m against logarithms of $(T - T_c)$, fig. 2b), d). The critical index β was found to be 0.75 ± 0.1 for 8CB and 0.76 ± 0.1 for 8OCB.

It appears from the experiment that, as the temperature is changed, the spherical tangentially anchored drops of nematic liquid crystal undergo a second-order paritybreaking phase transitions. These transitions occur due to the changes of the elastic constants $K_{\rm u}$ with temperature. Two states of the drop (untwisted and twisted) differ only with respect to the structural symmetry, neither phase state of nematic itself nor boundary conditions. Transitions between these states are similar to usual second-order transitions. The twisted bipolar structure behaves like an optically active object and leads to waveguide effect, in spite of nonactivity of nematic and isotropic matrix themselves.

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